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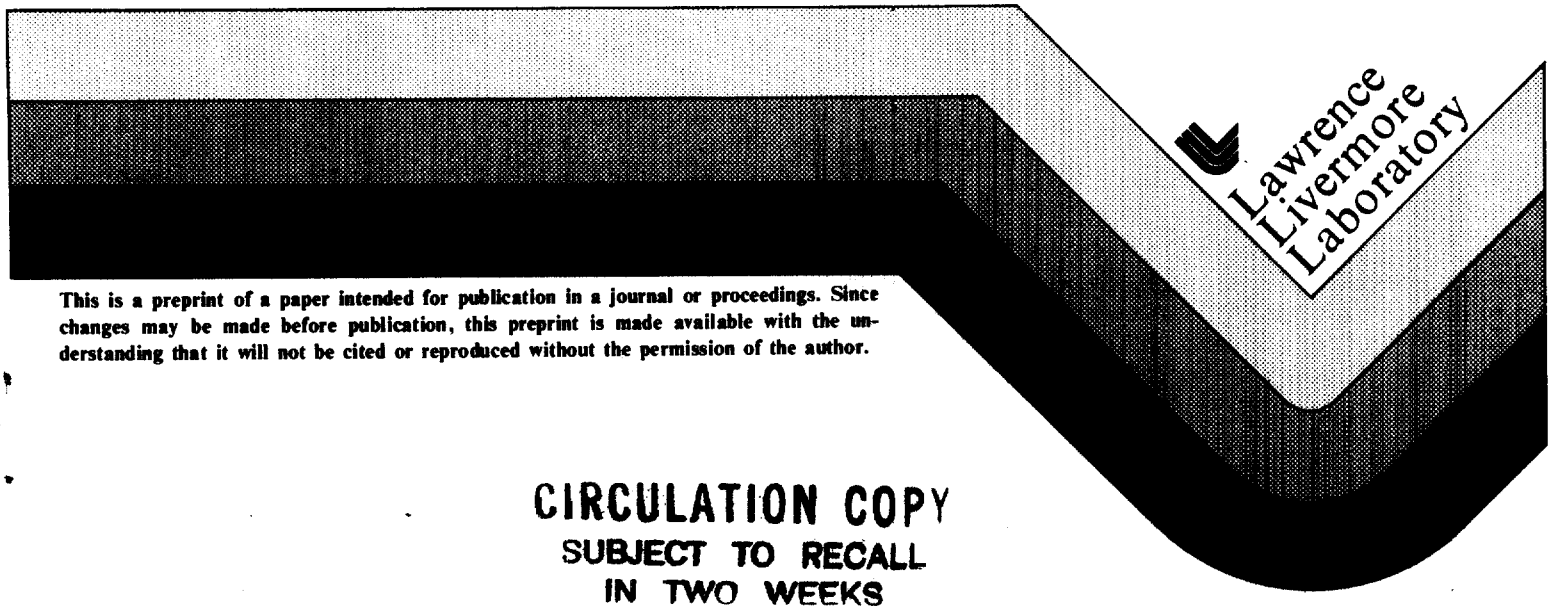

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The Influence of Propagation on Off-Resonant Molecular Excitation*

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ABSTRACT: We show that the proposal of Makarov, Cantrell, and Louisell regarding stimulated inelastic resonance fluorescence (SIRF) does not accurately represent the effects of propagation on off-resonant excitation of two-level model molecular absorbers.

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In a recent letter[1], Makarov, Cantrell, and Louisell (MCL) have discussed optical pulse propagation in connection with multiple-photon excitation of polyatomic molecules. They suggest, on the basis of an approximate analytic integration of the appropriate Maxwell-Bloch equations, for a two-level absorber that stimulated inelastic resonance flurorescence (SIRF) may be responsible for significant population transfer between the molecular ground level and all of the levels in the first excited vibrational state, particularly those excited levels that are far off resonance.

We have recently developed accurate computer codes for the solution of problems of light propagation[2] in multi-level media. We have adapted these codes to the SIRF problem in order to extend the semi-quantitative analysis of MCL, and we have come to different conclusions. We find that there is apparently no realistic off-resonant pumping pulse that will act in the manner described by the MCL two-level approximate solutions.

Our approach has been to use numerical methods to solve the propagation problem, given a specific input pulse shape. We have calculated the population transferred to an off-resonant excited level as a function of propagation distance in the absorbing medium. Over distances as much as twice the distance estimated by MCL to lead to complete inversion we find no more than five percent of the population in the excited level. This conclusion appears to be valid[3] for a wide range of input pulse shapes, from a very smooth gaussian, through a sequence of steeper hyper-gaussians, to a pulse that is perfectly square.

It was basically a qualitative spectral analysis that motivated the MCL discussion of this problem. Even if discussion of propagation can be seen to be too simplistic, the MCL suggestion that a resonant sideband pulse should begin to grow during propagation is reasonable. We have also studied this aspect of the problem by computing numerically the power spectrum of the reaction field as a function of propagation distance for bothf the hyper-gaussian and the square pulses. These power spectra are given in Figs. 1a and 1b.

Fig. 1a shows that the hyper-gaussian pulse generates no sideband reaction field strong enough to appear in the spectrum. Fig. 1b shows that the square pulse, however, does produce a sideband reaction field. Near to the entry plane the strengths of the central and sideband reaction fields are very nearly equal, as predicted by MCL.

However, significant departures from the MCL predictions are found as the pulses propagate away from the entry plane. This is not only true of the "realistic" square (hyper-gaussian) pulse but of the ideally square pulse as well. The power spectra of Figs. 1a and 1b make this clear. Although the sideband component in Fig. 1b is initially equal to the central component, it quickly becomes less important as propagation continues.

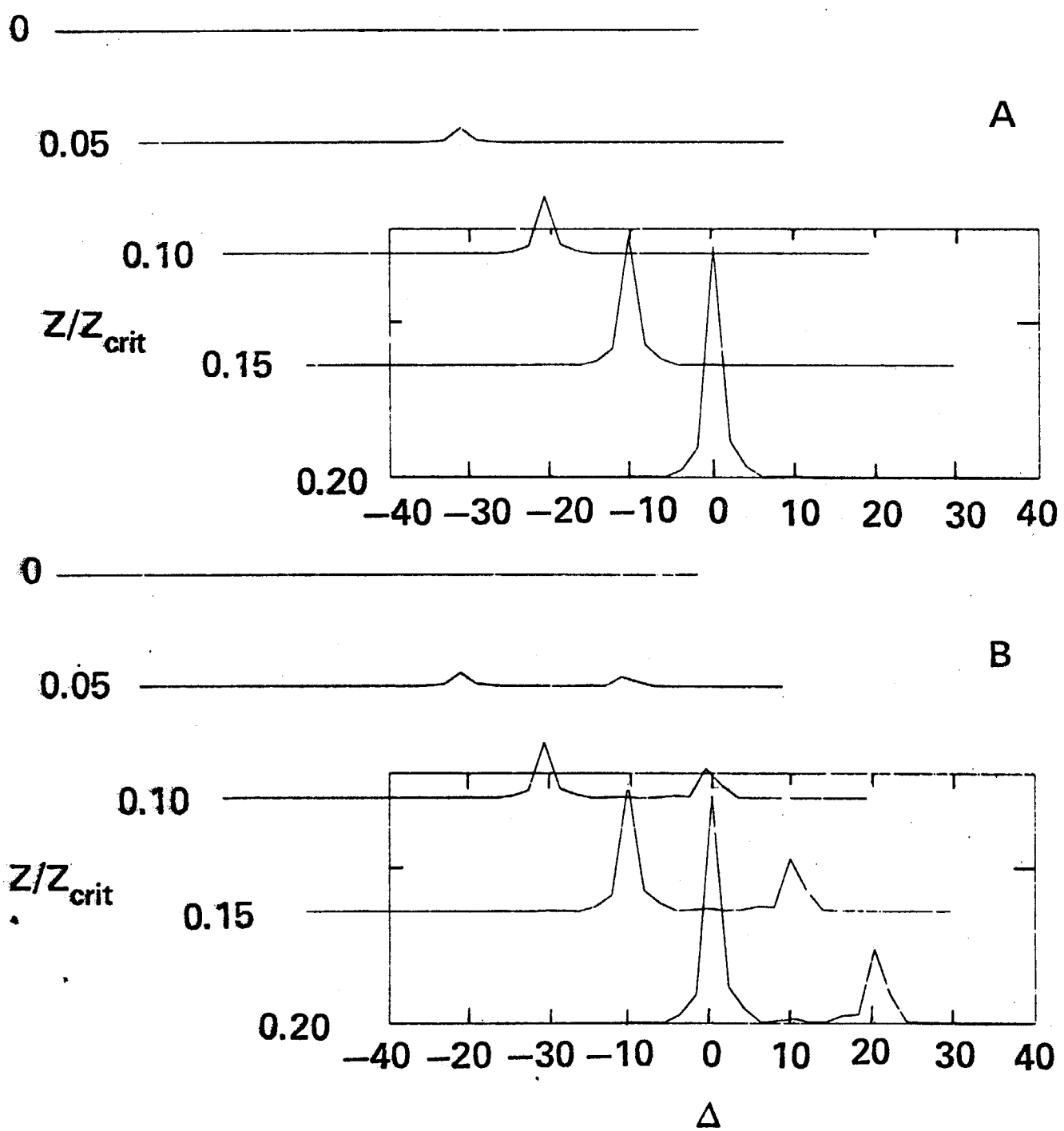
References

1. A. A. Makarov, C. D. Cantrell and W. H. Louisell, Optics Comm. 31, 31-36 (1979).

2. M. J. Konopnicki and J. H. Eberly, in Proc. of Tenth Annual Modeling and Simulation Conf., W. G. Vogt and M. H. Mickle, editors (Instrument Society of America, Pittsburgh, 1979). p. 1199. The numerical integration of coupled Maxwell-Bloch equations was initiated in the self-induced transparency studies of S. L. McCall and E. L. Hahn, Phys. Rev. 183, 457 (1969). See also A. Icsevgi and W. E. Lamb, Jr., Phys. Rev. 185, 517 (1969), and Chap. 4 of Ref. 3, below.

3. See, however, C. D. Cantrell, W. H. Louisell, and A. A. Makarov, in Advances in Chemical Physics, edited by J. Jortner and R. Levine (Wiley, New York, 1980), where MCL argue that a four-level model molecule may exhibit the effects proposed in Ref. 1.

Fig. 1. Spatial evolution of the reaction field energy spectrum. The spectrum is defined to be the absolute square of the Fourier transform of the difference between the electric field at the position Z and at the entry plane Z = 0 in the molecular absorber. Parts (a) and (b) show the hyper-gaussian and the exactly square cases, respectively. In both (a) and (b) the pulse and molecular parameters are the same as those used in Figs. 2 and 3. Here, however, we display results in the planes Z/Z_{crit} = 0, 0.05, 0.10, 0.15, 0.20, to emphasize how rapidly the entry-plane approximation of MCL become invalid.


0
A
0.05
0.10
Z/Z_{crit}
0.15
0.20
−40 −30 −20 −10 0 10 20 30 40
0
B
0.05
0.10
Z/Z_{crit}
0.15
0.20
−40 −20 −30 −10 0 10 20 30 40
Δ